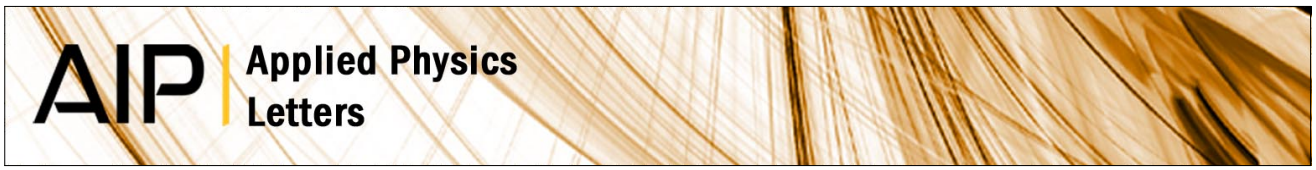

Growth temperature dependence of longrange alloy order and magnetic properties of epitaxial FexPt1−x (x0.5) films

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Growth temperature dependence of long-range alloy order and magnetic properties of epitaxial Fe_xPt_{1-x} ($x \simeq 0.5$) films

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The structural and magnetic properties of 1000-Å-thick epitaxial $Fe_xPt_{1-x}(001)$ alloy films, grown by molecular beam epitaxy with $x \simeq 0.5$, have been studied as a function of growth temperature. X-ray diffraction analysis showed that the long-range order parameter increased from near zero for films grown at 100 °C to a maximum of 0.93 in films grown at 500 °C. Over this range the magnetic easy axis changed from in-plane to perpendicular and the polar Kerr rotation increased strongly. Spontaneous long-range ordering in these films has an activation energy ~0.2 eV, consistent with an energy barrier for surface and step down diffusion. © *1996 American Institute of Physics.* [S0003-6951(96)03034-3]

The magnetic, magneto-optical and structural properties of intermetallic alloy films of Co_xPt_{1-x} and Fe_xPt_{1-x}, near the equiatomic composition $x=0.5$, are presently generating interest from both applied and basic viewpoints. In the search for longitudinal magnetic recording media with high magnetic anisotropy, high coercivity and low noise, these alloys have recently shown promising characteristics.[1] One essential feature is that they can undergo long-range chemical ordering to the $L1_0$ (CuAu I) phase[2] which comprises alternating atomic planes of Co(Fe) and Pt along the c-axis. Fully ordered FePt is predicted[3] to have one of the largest magnetic anisotropy energies ($\sim 1.6\times 10^8$ erg/cm^3) of any magnetic material and indeed, anisotropy energies of $K_u > 10^8$/cm^3 have been found experimentally in MBE-grown FePt films.[4,5] In the work of Coffey *et al.*,[1] the hard magnetic properties of magnetron-sputtered Co_xPt_{1-x} and Fe_xPt_{1-x} films were achieved by extended postgrowth annealing at temperatures up to 700 °C. This method has disadvantages as a manufacturing process because such high temperatures can cause irreversible changes in substrates for magnetic media. In this letter we demonstrate that long-range chemical ordering can be achieved spontaneously by coevaporation of Fe and Pt at temperatures ~300–500 °C, which are more amenable to manufacturing. We also correlate the magnetic properties (polar Kerr rotation and perpendicular magnetic anisotropy) with the long-range chemical order and, from a consideration of the ordering kinetics, we suggest the facile ordering results from surface diffusion.

The films were grown by coevaporation from e-gun sources for both Fe and Pt.[4,5] Film growth rates of Pt and FePt were ~0.1 and 0.2 Å/s, respectively. The films, ≃1000 Å thick, were grown on 150 or 7-Å-thick Pt(001) seed films on MgO(001) substrates with parallel epitaxy, as described earlier.[4,5] Film growth was characterized, *in situ* using both RHEED and LEED. Following growth, the films were characterized by x-ray diffraction, vibrating sample and torque magnetometry, and magneto-optical Kerr studies.

Specular x-ray diffraction scans for FePt films grown onto ≃150-Å-thick seed films at growth temperatures of 100, 200, and 550 °C are shown in Fig. 1(a). The progressive increase in the (001) and (003) peaks with increasing growth temperature is evident. These are superstructure peaks indicative of the presence of the long-range ordered $L1_0$ phase in the film. The long-range order is quantified[4,6] by the order parameter

$$S=(r_{\mathrm{Fe}}-x_{\mathrm{Fe}})/y_{\mathrm{Pt}}=(r_{\mathrm{Pt}}-x_{\mathrm{Pt}})/y_{\mathrm{Fe}}, \tag{1}$$

where $x_{\mathrm{Fe(Pt)}}$ is the atomic fraction of Fe(Pt) in the sample, $y_{\mathrm{Fe(Pt)}}$ the fraction of Fe(Pt) sites, and $r_{\mathrm{Fe(Pt)}}$ the fraction of Fe(Pt) sites occupied by the correct atom.

S was calculated by integrating intensities for the (001), (002), and (003) peaks extracted from fits to x-ray data. This procedure is described in detail elsewhere;[7] here we point out that the mosaic spread of each of the peaks was included in the calculation. Also included were an experimental Debye–Waller parameter ($\sigma=0.136$ Å), Lorentz polarization factor for both sample and monochromator, finite absorption in the film, and a sin θ factor to correct for the illuminated sample area. Figure 2 shows the long-range order parameter as a function of growth temperature. S was calculated from both (001)/(002) and (003)/(002) intensity ratios. The agreement between these values was within 4% at all temperatures[7] and the mean values are plotted in Fig. 2. Films with a departure of Δ in composition from $x=0.5$ can reach a maximum value of $S_{\max}=1-2\Delta$. Since the values of Δ (all <0.09) differ

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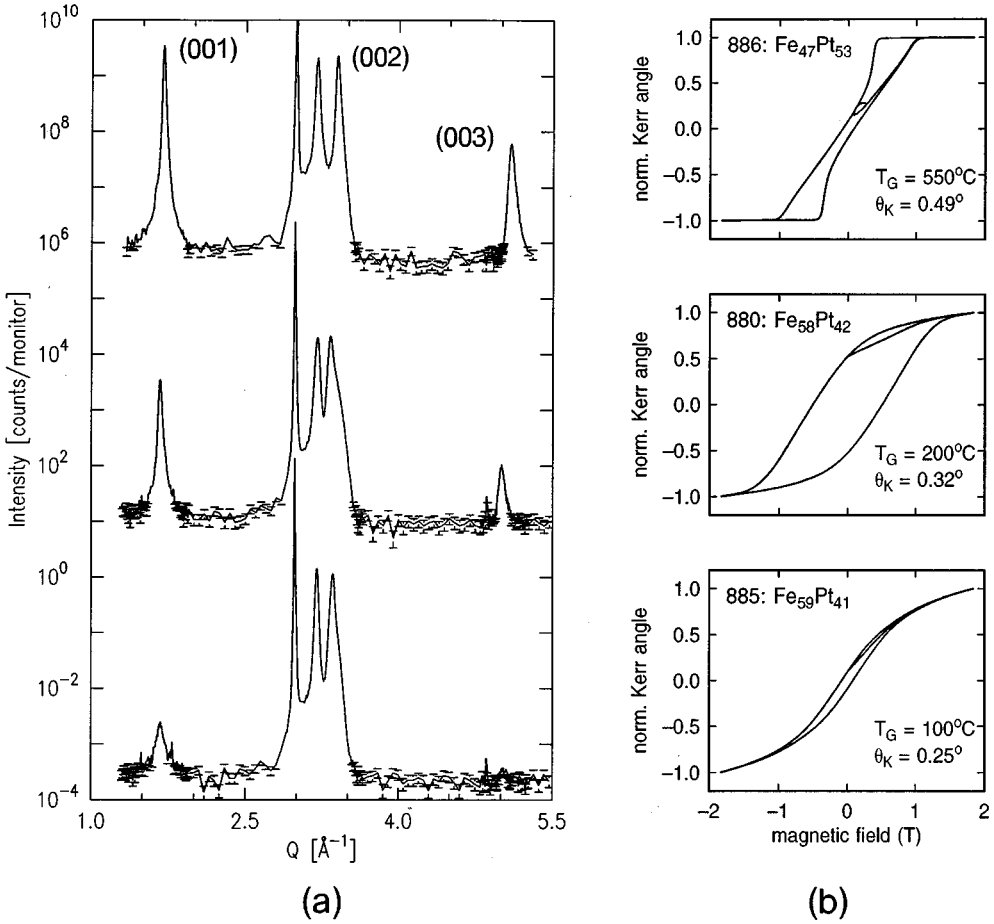

FIG. 1. (a) Specular ($\theta-2\theta$) x-ray scans for FePt films grown at 100, 200, and 550 °C on 150-Å thick seed films of Pt(001)/MgO(001). The intensities are plotted on a logarithmic scale vs the scattering vector $\mathbf{Q}=(4\pi \sin\theta)/\lambda$. The lower scan is for the film grown at 100 °C and the other scans have been offset for clarity. Note the progressive increase in FePt(001) and (003) intensities with growth temperature. The peaks at $\simeq$3.0 and 3.2 Å^{-1} are the MgO(002) and Pt(002) Bragg peaks from substrate and seed film, respectively. (b) Polar Kerr loops from the corresponding films.

slightly from film to film, the normalized values, $SS/S_{\max}$, are the relevant measure of the chemical order and are plotted in Fig. 2. This normalization removes any systematic errors caused by sample composition differences. Data for FePt films grown on thin seed films are shown by triangles and those for thick films by circles. At the temperatures 300 and 500 °C, where these data can be compared, there is no significant difference. S increases smoothly, peaking near 500 °C then falling slightly.

Partially ordered films grown in the temperature range 100–300 °C show no evidence of a first order (nucleation and growth) reaction which would be indicated by splitting of the (002) Bragg peak into distinct ordered and disordered components. Rather, the (002) peak is broad and asymmetric in this range, consistent with a dispersion of partially ordered domains in the film. This shows that the disorder–order transformation in the present films is of a continuous type rather than the first-order-type observed in annealed FePt films by Barmak *et al.*[8] One can estimate from the data of Fig. 2 the activation energy for spontaneous ordering. Since bulk diffusion is so slow below 600 °C, we assume that the ordering occurs only while the surface diffusion is operative, i.e., during some characteristic time that is independent of temperature and related to the inverse of the deposition rate. Then using Eq. (4) of Nowick and Weisberg[9] and noting that the growth temperature is much less than the order–disorder temperature of 1300 °C,[2] one can show that for reasonably small $S(\lesssim 0.5)$, $S(T)$ should exhibit an Arrhenius behavior with an activation energy that is a diffusional barrier height. Indeed, a plot of ln (S) vs $1/T$ has a linear region from 100 to 300 °C with a barrier height of 0.2 ± 0.04 eV. In the temperature regime where S exceeds 0.5 and saturates near unity, Eq. (4) of Nowick and Weisberg can also be used to deduce an expression for $S(T)$ that contains the diffusional barrier height. Analysis of our data ($T>220$ °C) using this formalism yields a barrier height of 0.18 ± 0.05 eV which is consistent with our analysis of $S(T)$ in the low-temperature regime. Interestingly, this energy is close to the energy barrier ($\simeq$0.2 eV) for adatom diffusion from a terrace to a lower terrace for Ag/Pt(111)[10] leading us to speculate that the rate limiting step for spontaneous FePt ordering is surface segregation of Pt and Fe onto adjacent terraces to form the $L1_0$ superlattice. We expect a similar process to operate in film growth by sputtering. The decrease in S above 500 °C may be due to a decrease in the equilibrium order parameter[11] or to the onset of surface induced disordering.[12] Further studies are needed to distinguish between these possibilities.

The magnetic and magneto-optical properties of our

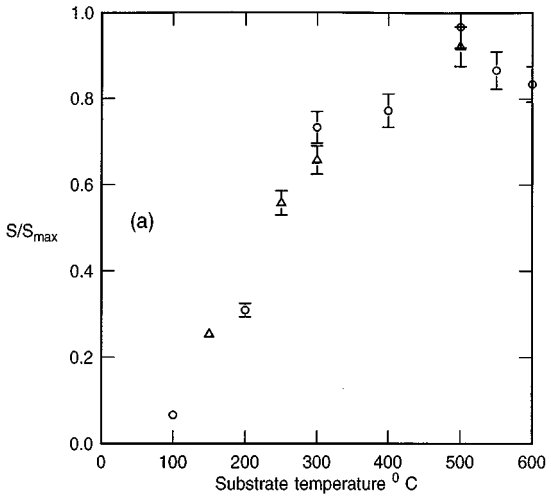

FIG. 2. Normalized long-range order parameter, $S/S_{\max}$, vs growth temperature.

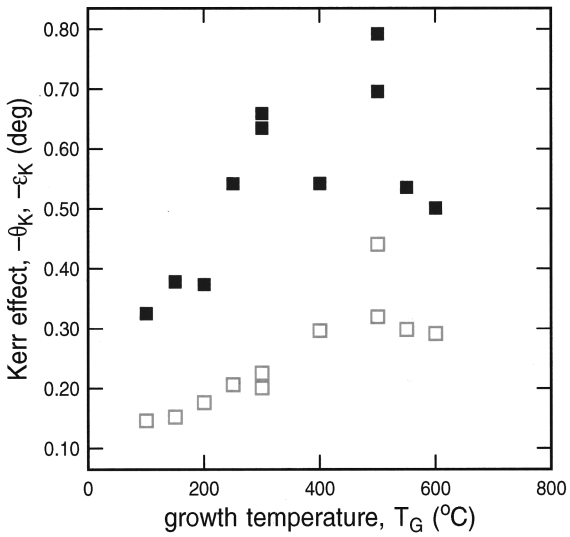

FIG. 3. Plot of magnitude of saturation polar Kerr rotation (θ_K) and ellipticity (ϵ_K) as a function of growth temperature. Wavelength 688 nm. Open squares: ϵ_K, closed squares: θ_K.

films correlate with the long-range order parameter. Figure 1(b) shows the polar Kerr loops for the samples of Fig. 1(a). Note that for the 100 °C film the perpendicular remanence is very small and the Kerr rotation (0.22°) is unsaturated at the maximum field available ($2T$). For the 200 °C sample the remanence is larger but the Kerr rotation (0.3°) remains unsaturated. However, for the 550° film the Kerr rotation is clearly saturated (0.5°) but the remanence remains small. These data and corresponding $M-H$ loops[14] show that the perpendicular magnetic anisotropy increases through this set of films. Remanence, however, is controlled by the magnetic domain structure and we have shown[5] that loops of the type seen for the 550 °C film are due to reverse-oriented magnetic domains presumably nucleated by structural defects in the film.

Qualitatively, the correlation between magneto-optical properties and long-range order is evident from a plot of saturation Kerr rotation and ellipticity as a function of growth temperature (Fig. 3). Errors in measurement of θ_K and ϵ_K are negligible compared with the size of the data points. The magnitude of these quantities increases with growth temperature, peaking at 500 °C then falling, as for S [see Fig. 2(a)]. Such a correlation is to be expected, since with increasing long-range order, the electronic structure of the film evolves from that of a chemically disordered cubic alloy to that of an atomically layered superlattice with uniaxial structural and magnetic symmetry. This correlation has been observed previously[14] in Co_3Pt alloy films. We also note that there is a strong dependence of the Kerr effect on the angle between the photon propagation direction and the c axis in FePt. This will be discussed in detail elsewhere.[15]

In summary, we have determined the dependence of long-range order on growth temperature during ultrahigh vacuum coevaporation of epitaxial FePt films on Pt/MgO(001). The order parameter increases from near zero at 100 °C to a peak of 0.93 ± 0.04 at 500 °C. Considerable ($S=0.74$) long-range order is present at 300 °C and the magnetic anisotropy is already appreciable at this temperature showing that it should be possible to achieve optimal magnetic anisotropy and coercivity by film growth at moderate substrate temperatures, far below those needed for post-growth annealing of films grown at room temperature. The activation energy for ordering is consistent with surface and step down diffusion as the rate limiting step.

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